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| Article Title: | | | | | | | | | | | | | | |
| Volume Number: | | Report Number: | | Pages: | | | | | | | | | | |
| Issue Number: | | Series Number: | | Year of Publication: | | | | | | | | | | |
| Publisher: | | | | | | | | | | | | | | |
| Remarks: | | <i>See attached: citations with yellow tab 472913</i> <i>(183)</i> | | | | | | | | | | | | |
| Monthly Accession Number: | | | | | | | | | | | | | | |
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| Page Count | | | <i>7</i> | | | | | | | | | | | |
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5876036 INSPEC Abstract Number: B9805-7260-012

Title: The application of poly(phenylene) type polymers and oligomers in electroluminescent color displays

Author(s): Tasch, S.; Brandstatter, C.; Graupner, W.; Hampel, S.; Hochfilzer, C.; List, J.W.E.; Meghdadi, F.; Leising, G.; Schlichting, P.; Rohr, U.; Geerts, Y.; Scherf, U.; Mullen, K.

Author Affiliation: Inst. für Festkörperphys., Tech. Univ. Graz, Austria

Conference Title: Flat Panel Display Materials III. Symposium p.325-30

Editor(s): Fulks, R.T.; Parsons, G.N.; Slobodin, D.E.; Yuzuriha, T.H.

Publisher: Mater. Res. Soc, Pittsburgh, PA, USA

Publication Date: 1997 Country of Publication: USA xi+338 pp.

ISBN: 1 55899 375 4 Material Identity Number: XX97-02853

Conference Title: Flat Panel Display Materials III. Symposium

Conference Date: 31 March-3 April 1997 Conference Location: San Francisco, CA, USA

Language: English

Subfile: B

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...Abstract: the oligomer hexaphenyl, are very suitable materials for realisation of efficient, stable, large area blue organic light emitting diodes (OLEDs). The emission of blue OLEDs can be efficiently converted into all other emission colours...

... EL device is covered with highly fluorescent dye/matrix layers, which are excited by the blue emission and emit lower energy photoluminescent light. Secondly, a new method for producing efficient white light emitting polymer diodes (e.g. for backlight sources in liquid crystal displays) based on...

...Identifiers: organic light emitting diodes ; ...

... OLED efficiency...

... OLED stability...

...blue OLED emission conversion

11/3,K/6 (Item 6 from file: 2)

DIALOG(R) File 2:INSPEC

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5696373 INSPEC Abstract Number: B9710-4260D-037

Title: White and unsaturated color organic light emitting diodes

Author(s): Dodabalapur, A.; Strukelj, M.; Jordan, R.H.; Rothberg, L.J.; Miller, T.M.

Author Affiliation: AT&T Bell Labs., Murray Hill, NJ, USA

Conference Title: Electrical, Optical, and Magnetic Properties of Organic Solid State Materials III. Symposium p.59-63

Editor(s): Jen, A.K.-Y.; Lee, C.Y.-C.; Dalton, L.R.; Rubner, M.F.; Wnek, G.E.; Chiang, L.Y.

Publisher: Mater. Res. Soc, Pittsburgh, PA, USA

Publication Date: 1996 Country of Publication: USA xvi+710 pp.

Material Identity Number: XX96-02015

Conference Title: Electrical, Optical, and Magnetic Properties of Organic Solid State Materials III. Symposium

Conference Date: 27 Nov.-1 Dec. 1995 Conference Location: Boston, MA, USA

Language: English

Subfile: B

Best Available Copy

**MATERIALS RESEARCH SOCIETY
SYMPOSIUM PROCEEDINGS VOLUME 471**

Flat Panel Display Materials III

Symposium held March 31-April 3, 1997, San Francisco, California, U.S.A.

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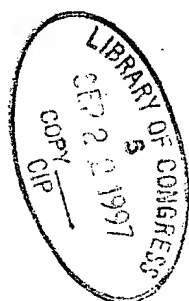
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PITTSBURGH, PENNSYLVANIA



TK7872
.L56 F5
1997

Preface

Materials Research

P.

***Deposition of Prec
Display Applicatio**
*R. Pethö, C. De
and N. Turner*

***Substrate Issues fo**
Dawne M. Moffe

**Flexible, Lightweig
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S.D. Theiss, C.C

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Constant Planariz
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*Je-Hsiung Lan,
and Jerzy Kanik*

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Displays**
*A.D. Williams, J
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***Invited Paper**

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University Microfilms Inc., 300 North Zeeb Road, Ann Arbor, Michigan 48106

CODEN: MRSPDH

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Published by:

Materials Research Society
506 Keystone Drive
Warrendale, PA 15086
Telephone (412) 779-3003
Fax (412) 779-8313
Website: <http://www.mrs.org/>

Library of Congress Cataloging in Publication Data

Flat panel display materials III : symposium held March 31-April 3, 1997,
San Francisco, California, U.S.A. / editors, Ronald T. Fuls,
Gregory N. Parsons, David E. Slobodin, Todd H. Yuzurha
p. cm.—(Materials Research Society symposium proceedings ; v. 471)
Includes bibliographical references and index.
ISBN 1-55899-375-4

1. Liquid crystal displays—Materials—Congresses. 2. Thin film
transistors—Materials—Congresses. 3. Information display systems—
Materials—Congresses. I. Fuls, Ronald T. II. Parsons, Gregory N.
III. Slobodin, David E. IV. Yuzurha, Todd H. V. Series: Materials
Research Society symposium proceedings ; v. 471.
TK7872.L56F5 1997
621.3815'422—dc21

Manufactured in the United States of America

97-22652
CIP

between the anode and cathode is about 700 μm . As shown in low for the first 6 sec. It may be due to slightly thin deposition on time increase, the higher brightness is obtained since the his time. The measured thickness of each phosphor is about 6-um thickness for the maximum brightness exceeds.

of the phosphor in a fully sealed FED environment, electron (10)(X500 μm) is measured after the sealing. The pixel is biased ote voltages of 250 volts. As shown in figure 8, there is no adjacent color lines, which results from no-cross contamination. The gap between the lines is also 100 μm .

osition conditions of each phosphor. Since the emission of to those of other colors, different thickness of each color is tics as shown in table 1.

| Material | Thickness(μm) | CIE(x) | CIE(y) |
|----------------------|----------------------------|--------|--------|
| 1S:Cu,Al | 5-6 | 0.311 | 0.55 |
| 2O ₂ S:Eu | 10 | 0.61 | 0.34 |
| 3S:Ag,Cl | 7 | 0.17 | 0.094 |

on conditions used for the FED device

as been successfully utilized to deposit primary low voltage for the development of 4-inch full color FED devices. The sphors, isopropyl alcohol with various concentration of nitrite of deposition time and applied voltages are investigated and chamber and in fully-sealed 4 inch FED environment. Even is under development to get optimum deposition conditions, nient to be applied to the FED device.

rey, and E.R. Westberg, *J. Appl. Phys.*, **47**, 5248 (1976). ra, and K. Yano, *ASIA Displays '95 TechDig*, 617 (1995). /MC'91, 6 (1991).

1 D.E. Sargent, *J. Electrochem. Soc.*, **117**, 1456 (1970). Aslam, *Adv. Electron. Phys.*, **22A**, 571 (1966).

zky, T. Avalos, and K.R. Hesse, *J. Electrochem. Soc.*, **137**, zky, T. Avalos, and K.R. Hesse, *J. Electrochem. Soc.*, **137**,

THE APPLICATION OF POLY(PHENYLENE) TYPE POLYMERS AND OLIGOMERS IN ELECTROLUMINESCENT COLOR DISPLAYS

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ABSTRACT

Due to their high photoluminescence efficiency (>30%), high environmental stability and the good charge transport properties the derivatives of poly(paraphenylene) (PPP), as the ladder-type PPP (LPPP) and the oligomer hexaphenyl, are very suitable materials to realise efficient, stable, large area blue organic light-emitting diodes (OLEDs). The emission of blue OLEDs can be efficiently converted into all other emission colors either by an external color conversion technique (ECCCT) or an internal color conversion technique (ICCT) and hence are very interesting for a number of display applications.

Firstly, we demonstrate the realisation of efficient red-green-blue (RGB) emission colors (representing the RGB-pixels in a multicolor display) by an external CCT. In this case the blue EL device is covered with highly fluorescent dye/matrix layers, which are excited by the blue emission and emit photoluminescence light in a lower energetic range.

Secondly, a new method for producing efficient white light-emitting polymer diodes (which are interesting for e.g. backlight sources in liquid crystal displays) based on a blend of two polymers is presented: a blue light-emitting m-LPPP and a red-orange emitter poly(phenylene-co-dithienylbenzene) (PPDB). The red-orange emission is created within the EL device (ICCT) by an excitation energy transfer from m-LPPP into the energetically lower lying states of PPDB. This internal excitation energy transfer is very efficient, so that only a concentration of 0.05 weight % PPDB in the polymer blend is required in order to obtain white light emission.

INTRODUCTION

Organic light-emitting diodes are based on the principle that electrons and holes are injected into the organic semiconductor from opposite electrodes, when an external bias (usually between 5 and 20 V) is applied [1-6]. The EL emission is observed, when the combination of oppositely charged carriers leads to the formation of excited states (singlet excitons), which then decay radiatively. OLEDs are very suitable materials for application in flat-color-panel-displays due to the high [7-9] achievable luminance (> 10000 cd/m^2 [10,11]), the high EL efficiency (up to 4% [12-13]) and the low driving voltages (<10 V) of organic thin film devices. We show how self-emissive displays and also efficient white OLEDs for usage in liquid crystal displays (LCD) can be produced based on blue OLEDs using external and internal color conversion techniques.

a. Color Flat Panel Displays based on OLEDs

The possibility of producing large area, flat devices with a high brightness (comparable to fluorescent tubes) makes the OLEDs promising candidates for the application in color flat panel

displays (CFPD). The realisation of color dots (pixels) building up a CFPD based on organic EL devices could be achieved via: (i) blue, green and red OLEDs, (ii) OLEDs tuneable from blue to red emission controlled by the voltage [12, 16, 17], current, local temperature or another device parameter, (iii) large area EL devices with a spectrally broad white emission, covered with red, green or blue absorption filters, (iv) blue large area EL devices covered with dye/matrix layers, which are excited by the blue light and subsequently emit photoluminescence light in a lower energetic range [18, 19].

In this article we focus on the last method, which is based on the principle of an external color conversion technique. This method, which uses blue OLEDs, allows to achieve bright multicolor emission in a very simple way and hence is one of the most promising techniques for realising CFPDs.

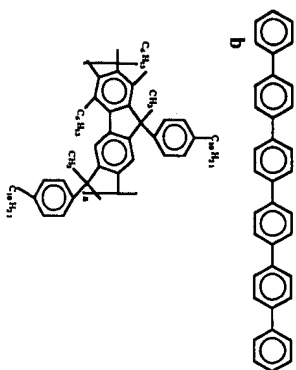


Figure 1: chemical structure of a Parahexaphenyl (PHP) and b methylated ladder-type poly(paraphenylene) (m-LPPP)

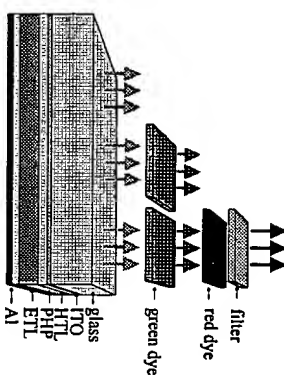


Figure 2: external CCT based on a blue OLED: glass/transparent electrode (ITO)/hole-transport-layer (HTL)/PHP/electron-transport-layer (ETL)/Al. The blue PHP emission light is converted by covering the PHP OLED with a green dye layer (→ green color) and a red dye layer together with a suitable dielectric filter (→ red color)

We present the realisation of bright red, green and blue emission colors by an all organic external CCT using an EL device of the organic semiconductor Parahexaphenyl (PHP) (Fig. 1) together with suitable matrix dye layers and dielectric filters.

The EL devices were produced in an air stable multilayer structure (s. Fig. 2) using Indium-Tin-Oxide (ITO) as the transparent bottom electrode (anode) and an oligoazomethine as a hole transport layer. The homogeneous polycrystalline PHP layers were deposited onto oligoazomethine via molecular beam deposition. Finally Diaminodifluorobiphenyl and Al were evaporated on top of PHP as electron-transport-layer and as the low work function contact, respectively.

The shape of the current/voltage characteristics of the PHP OLEDs is strongly asymmetric. In forward direction (ITO as the anode), the onset of the current and hence the EL occurs at an electric field of 0.3 MV/cm (corresponds to 5V bias voltage), whereas in reverse direction only a small leakage current below 1 μ A passes through the device. Under cw-operation luminance values of around 360 cd/m^2 and the external EL quantum efficiencies between 1% and 2% are observed.

A fundamental advantage of the deep blue PHP emission light (CIE coordinates $x = 0.15$, $y = 0.06$ [20]) is that it can be converted into any other visible color, e.g. with the external CCT introduced above, since the blue emission is located at the high energy side of the visible spectrum. To obtain a certain emission color the light can be spectrally purified (with a minimal

intensity loss) by using a suitable dielectric band pass filter. Such interference filters were produced by subsequent evaporation of high and low refractive index materials TiO_2 and MgF_2 of defined thickness.

Applying this all organicECCCT emission colors (as green, yellow, orange, magenta and red) throughout the whole visible range were produced by covering the EL devices with dye layers of different concentration, and if necessary, with suitable dielectric filters. The internal quantum efficiency for this ECCCT is above 90%.

Regarding to the realisation of CFPD the efficient production of the two other basic colors (besides blue) green and red is of great interest. With this all organic color conversion technique RGB-emission colors with CIE coordinates (green: $x = 0.27$, $y = 0.62$; red: $x = 0.65$, $y = 0.34$) similar to the basic colors which are used in commercial color TVs were produced (s. Fig. 3) [19].

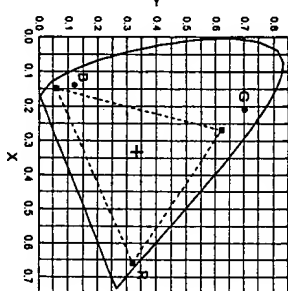


Figure 3: CIE-graph containing the CIE coordinates of the produced RGB-colors (squares) compared to RGB-colors used in cathode ray tubes (circles) (the dotted line marks all colors, which can be produced by the RGB colors)

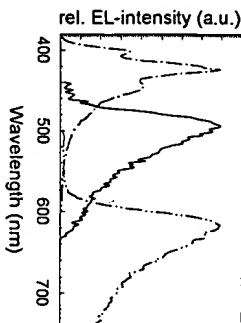


Figure 4: normalized spectra of the electroluminescence of the PHP OLED (---), the green (—) and the red (— · —) emission produced by ECCCT.

The green and red light emission is produced as follows:

bright green emission is obtained by using a layer of the dye Coumarin 102 in poly(methylmethacrylate) (PMMA) of a high optical density (>3). The Coumarin 102 is a very suitable dye for the color conversion due to the strong overlap between the Coumarin absorption and the PHP EL emission spectrum and due to its efficient green PL light emission. The absolute external conversion efficiency from the blue PHP EL emission into the green was recorded to be around 40% [19]. The green emission (luminance $\approx 1100 \text{ cd/m}^2$) is around three times brighter to the human eye than the blue pumping light from the PHP (360 cd/m^2), which is due to the fact the human eye sensitivity is much higher in the green spectral range than in blue (s. Fig. 3, 4).

The red emission, which is more difficult to achieve, can be obtained by covering the Coumarin/PMMA layer with a layer of the red dye Lumogen F300 in PMMA. This red dye strongly absorbs in the green spectral range and therefore converts the green excitation into red emission via PL emission. To obtain an emission color, which is located in the pure red spectral region ($x = 0.65$, $y = 0.34$ - s. Fig. 3) the spectrum can be purified with a band pass filter (s. Fig. 2, 4). The conversion efficiency from the blue (via the green) to the red was determined to be 10% [19]. The observed intensity losses are mainly due to waveguiding within the red dye layer, but also due to reflection losses at the filter.

The external conversion efficiency can be significantly increased, when the index matching of the dye/matrix layers to the EL devices is improved, so that waveguiding losses are reduced. When the small airgap between the color conversion films and the EL device are eliminated, e.g. by depositing the color-conversion layer directly onto the device, the quantum efficiencies for color conversion from the blue into the green of around 90% and for the blue into the red of around 80% are obtained [21].

b. Efficient white OLEDs for LCDs

The production of white OLEDs is difficult, since no single active material is known up to now, which produces white light (although the emission of most of the light-emitting organic semiconductors is spectrally very broad). There have been several concepts presented in order to achieve white light emission with OLEDs based on thermally vacuum deposited organic metal complexes:

- (i) using an OLED consisting of two or more active layers, where the emission is a superposition of the EL emission spectra of these light-emitting layers [22-27]. The recombination zones in these OLEDs are spatially controlled by the application of hole blocking layers [23-25].
- (ii) white light obtained by spectral filtering of the emission light of a suitable broad emitter [28].
- (iii) white light can be produced by the superposition of red-green and blue OLEDs. RGB-LEDs based on organic materials have been presented in Ref. [18,19,29].

We present the realization of stable and highly efficient white polymer LEDs based on an ICT in a blend of two polymers: a blue-light emitting ladder-type (polyparaphenylene) (m-LPPP) (s. Fig. 1) and a new red light emitting poly(perylene-co-diethynylbenzene) (PPDB) (for synthesis see Ref. [30,31]). The spectrally broad blue m-LPPP emission spectrum (CIE coordinates $x=0.24, y=0.39$) in combination with only one red-orange light emitting component (PPDB) ($x=0.61, y=0.39$) in a polymer blend enables us to realize white light emission (s. Fig. 5, 6) [32]. In contrast to the various concepts presented above, the red-orange component in the emission spectrum is generated by an excitation energy transfer from the m-LPPP polymer chains into the energetically preferred states of PPDB [33]. One important technological advantage of these efficient white OLEDs compared to other LEDs, is the ease of their production by a solution casting process of the active layer.

According to colorimetry, the coordinates of a color, which is produced by mixture of two colors, can be found on the connecting line between the coordinates of the two colors in the CIE graph (s. Fig. 5), where the exact position is determined by the quantitative composition. It is obvious from the position of the CIE coordinates of the m-LPPP and PPDB emission, that a mixture of m-LPPP and PPDB emission colors can produce white light emission near the equilibrium white point ($x=1/3, y=1/3$). In OLEDs various color compositions can be realized, when the active layer is built up by a blend of different light emitting polymers [26,27]. In the m-LPPP/PPDB blend a very efficient transfer of electrons, holes or entire excitations from m-LPPP into the energetically lower lying PPDB-states occurs. For PPDB concentrations above 3 weight% the emission stems exclusively from PPDB and the m-LPPP emission component vanishes [33].

We obtained a white OLED, when using a 80 nm thick active layer of a m-LPPP/PPDB polymer blend containing 0.05 weight% PPDB. These OLEDs emit stable white light to the human eye *independent* on the bias voltage. However, in the EL spectra an increase of the emission coming from PPDB can be observed, when the applied external electric field is

exceeding values of 2.5 MV/cm. The external EL quantum efficiency of the white OLED is around 0.8%.

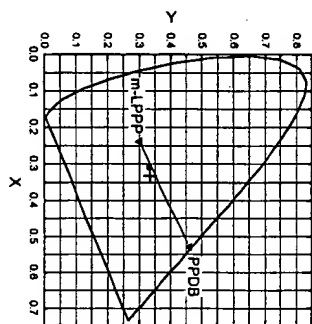


Figure 5: CIE coordinates of the EL of m-LPPP ($x=0.31, y=0.33$), the PL of a PPDB ($x=0.53, y=0.46$) and of the white LEDs operated at 30V ($x=0.31, y=0.33$).

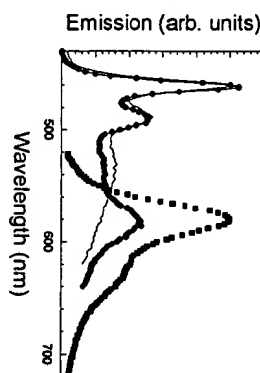


Figure 6: EL (—) spectrum of m-LPPP ($d_{\text{LPPP}}=100$ nm), PL spectrum of PPDB-solution (squares) compared to EL spectra of the white OLED ITOM-LPPP/PPDB/Al (thickness of active polymer layer is 80 nm) recorded at 30V (circles)

We further increase the efficiency of the OLED without losing the advantage of the simple processing of the OLED by introducing poly(methylmethacrylate) (PMMA) in the polymer blend, which acts as a charge carrier-blocking material (instead of using suitable electron- or hole transport layers, which require a thermal evaporation). In this case the efficient energy transfer from the blue to the red is partly hindered by PMMA, so that the concentration of PPDB in the blend has to be increased in order to produce white OLEDs. For a polymer blend containing around 10 weight% PMMA, the PPDB concentration has to be 0.08% to obtain pure white light emission (CIE coordinates $x=0.31, y=0.33$) (s. Fig. 5, 6). The IV characteristics of the white OLEDs containing PMMA reveal two important advantages compared to those without PMMA: (i) the current flow below the threshold electric field (*leakage current*) is reduced to values in the 10^{-8} A-range (ii) the threshold electric field is significantly lowered and occurs at values below 1 MV/cm [32]. The maximum external EL quantum efficiency of this white OLED containing PMMA is around 1.2 %. There is no significant dependence of the EL spectra of the white OLED containing PMMA on the applied bias voltage.

OLEDs are very promising for future application in various types of displays. Blue OLEDs are of special interest, since their emission can be converted into any other visible emission color using either an external or an internal CCT. We have demonstrated the realisation of bright (up to 3900 cd/m²) RGB-emission colors by an ECCT and the realisation of efficient white OLEDs (up to 1.2 % photons/electron) by an ICT using the blue light emitting materials m-LPPP and PHP.

ACKNOWLEDGEMENTS:

We would like to thank Dr. Koller (Lambda) for providing us the pure dye systems for the ECCT experiments. The financial support of this research work by the Spezialforschungsbereich 'Elektroaktive Stoffe' and the Jubiläumsfond der Österreichischen Nationalbank is gratefully acknowledged.

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LUMINESCENT STUDIES SILICA AEROGELS FOR

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ABSTRACT

The remarkable optical and establish their utility as unique, m luminescent materials for display, photoluminescence, absorption, at aerogels and aerogels doped with Eu

INTRODUCTION

Aerogels represent a novel cl electronic, thermal, and structural p in which luminescent materials are i these properties include low wei chromophore, and physical robustn roughly be 99.8% air and 0.2% mat made up of clusters of materials whi and extensive internal surface area robust, able to support 1600 times t doped with a variety of atoms and n In addition, silica aerogels are non-h We studied the optical prof Eu³⁺, rhodamine 6G, and fluorescei photoluminescence excitation (P) performance for FPD purposes. W those observed for each chromoph. Our results demonstrate the potentia dyes and other luminescent materi.

EXPERIMENT

The preparation of the aerog Transmission and absorption Optical excitation for PL studies wa mode-locked Ti:sapphire laser was excitation pulses varying from 355- from a Ti:sapphire regenerative amp at a 1 KHz repetition rate, and (3) recorded with a fluorimeter and als optical multichannel analyzer. Th optical system. Photoluminescence

RESULTS

Figure 1 shows the PL spec This spectrum can be compared wi the chromophore-doped silica aero;

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